

Ground State of a Spin System with Two- and Four-spin Exchange Interactions on the Triangular Lattice

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We study a spin system with both two- and four-spin exchange interactions on the triangular lattice as a possible model for the nuclear magnetism of solid ^3He layers adsorbed on grafoil. The ground state is analyzed by the use of the mean-field approximation. It is shown that the four-sublattice state is favored by introduction of the four-spin exchange interaction. A possible phase transition at a finite temperature into a phase with the scalar chirality is predicted. Application of a magnetic field is shown to cause a variety of phase transitions.

It is a great pleasure to dedicate this paper to Professor Wolfgang Götze for his sixtieth birthday, in whose research group at Max Planck Institut für Physik und Astrophysik and Die Technische Universität München one of the authors (K.K.) had the privilege to spend very pleasant and profitable years as a postdoctoral research associate, and to wish him many more happy years of activity in theoretical physics.

I. INTRODUCTION

Nuclear magnetism of the two-dimensional ^3He layers adsorbed on a grafoil has been a puzzle since Greywall and Busch reported the specific heat data at the coverage $\rho = 0.178 \text{ \AA}^{-2}$.^{1,2} At this coverage the adsorbed ^3He atoms form two layers and each layer solidifies into a triangular lattice. The first layer is a high-density solid and is considered not to contribute to the magnetism in the mK temperature region. The specific heat data down to 2 mK showed a prominent peak at 2.5 mK as is expected from the conventional Heisenberg antiferromagnet (HAF) on the triangular lattice. But integration of their data extrapolated linearly to lower temperatures indicated that a half of the total magnetic entropy will not be released down to the absolute zero temperature. Elser proposed that the second layer forms a $\sqrt{7} \times \sqrt{7}$ structure in registry with the first layer triangular lattice.³ Then it decomposes into inequivalent A and B sites which may be magnetically decoupled. Since A sites form a kagomé lattice, he proposed that the missing entropy may be explained by the HAF on the kagomé lattice rather than that on the triangular lattice. Inspired by this proposal many investigations were accomplished on the HAF on the kagomé lattice in recent years.⁴ According to numerical studies of finite clusters the kagomé HAF shows a

second peak in the specific heat at a low temperature, which might compensate the missing entropy.^{3,5} On the other hand, Roger showed for a cluster with 16 spins that a second peak can be also produced by effects of the multi-spin interactions which should be of considerable amount in solid ^3He systems.⁶

Recent specific heat data down to 0.1mK at the coverage $\rho = 0.180 \text{ \AA}^{-2}$, however, do not exhibit any sharp second peak in the temperature region where the kagomé HAF predicts.⁷ Furthermore the estimate of the entropy released at temperatures above 0.1mK nearly exhausts the total magnetic entropy of the second layer. The peak at $T \simeq 2.5 \text{ mK}$ found by Greywall and Busch turned out to have a much broader tail in the low temperature region than was first thought. It is clear that these features of recently obtained specific heat data cannot be explained in terms of either the HAF on the triangular lattice with nearest neighbor exchange couplings or that on the kagomé lattice. So the behavior of the specific heat remains as a puzzle though there is not missing entropy any longer.

It is well known that magnetic interactions due to ring exchanges involving more than two spins are important in solid ^3He . The famous *uudd* state in bcc solid ^3He is incorporated by the four-spin exchange interaction.⁸ The exchange frequencies in bcc solid ^3He computed by using the path-integral Monte Carlo method agree fairly well with estimates from experiments.⁹ For a monolayer of ^3He at the coverage $\rho = 0.0785 \text{ \AA}^{-2}$ on grafoil the cyclic-exchange frequencies were computed by Bernu et al. in a similar way.¹⁰ Their result shows that the three-spin exchange term predominates the others at this coverage and the high-temperature specific heat is governed by the two-, three- and four-spin exchange interactions while the six-spin exchange term as well is necessary to be taken into account to estimate the Weiss temperature properly.

The recent experimental results indicate that we have to take account of all spins in the second layer in order to understand its magnetism. We assume that the difference between A and B sites causes negligible effects on the translational symmetry of the magnetic interactions and that the magnetism of the second layer may be described by an $S = 1/2$ spin system on the regular triangular lattice. We employ the Hamiltonian with the two-, three- and four-spin exchange interactions as an effective Hamiltonian for the second layer of adsorbed ^3He , though no estimates of exchange frequencies were reported for the second layer so far. This model may describe the ^3He monolayer adsorbed on plated graphite as

well.^{11,12} Since three-spin exchange terms map into usual two-spin exchange couplings, the model is expressed by the two-parameter Hamiltonian

$$H = J \sum_{\langle i,j \rangle}^{\text{n.n.}} \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + K \sum_p h_p, \quad (1)$$

where $\boldsymbol{\sigma}_i/2$ is the nuclear spin of the ${}^3\text{He}$ atom at the site i and \sum_p denotes the sum over all possible diamonds composed of two nearest neighbor unit triangles. The four-spin exchange term of the diamond p depicted in Fig. 1(a) reads

$$h_p = \sum_{\langle \alpha, \beta \rangle} \boldsymbol{\sigma}_\alpha \cdot \boldsymbol{\sigma}_\beta + (\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_b)(\boldsymbol{\sigma}_c \cdot \boldsymbol{\sigma}_d) + (\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_d)(\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_c) - (\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_c)(\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_d), \quad (2)$$

where $\sum_{\langle \alpha, \beta \rangle}$ denotes the sum over all possible pairs of four spins on the diamond. The parameter J and K are related to the exchange frequency J_n of the n -body cycle of the nearest neighbors as

$$J = -J_2/2 + J_3 \quad (3)$$

and

$$K = -J_4/4. \quad (4)$$

It should be noted that the exchange frequency J_n is always negative for any n .¹³ The magnitude of the two-body exchange frequency J_2 is expected to be large at a low enough atomic density of the layer but it decreases with density more rapidly than J_3 .^{8,6} Thus the parameter J is expected to be positive at a low density while it should be negative in the high density region. On the other hand K is always positive. We expect that a wide parameter region of the ratio of J to K may be realized by varying the coverage of ${}^3\text{He}$ layers in a realistic system. It is therefore useful to investigate what kind of the ground state occurs in the system described by the Hamiltonian (1). Calculations of the specific heat and the susceptibility of a 16-spin cluster were reported by Bernu et al. previously.¹⁰

II. THE GROUND STATE IN THE CLASSICAL LIMIT

As a first step to study the spin system described by the Hamiltonian (1), we examine the ground state in the classical limit. Since we neglect quantum fluctuations, we treat $\boldsymbol{\sigma}_i$ as a unit classical vector in the following. When $K = 0$ the ground state of the system is well known, that is, the ferromagnetic state for $J < 0$ and the state with the so-called 120° structure for $J > 0$. On the other hand we cannot guess the ground state of the system intuitively when $J = 0$, since the Hamiltonian is governed by the unusual four-spin term. So we use a variational

argument. We divide the total Hamiltonian into a sum of those of interpenetrating hexagons with 7 spins. That is

$$H = \frac{K}{2} \sum_h H_h, \quad (5)$$

where H_h is the Hamiltonian of the hexagon depicted in Fig. 1(b) and is expressed as a sum of Hamiltonians (2) for six diamonds contained in the hexagon. Then we searched numerically for the spin configuration which minimizes H_h . It has turned out that the lowest-energy spin configuration is a four-sublattice spin structure where any two spins on different sublattices make the relative angle α where $\cos \alpha = -1/3$ (see Fig. 2(b)). We can divide the whole lattice into four sublattices such that four vertices of every diamond belong to different sublattices. The spin configuration which minimizes a hexagon, therefore, can be extended to the whole lattice in such a way that the state has the lowest energy for all hexagons in eq. (5) consistently and so realizes the ground state of the whole lattice. The spin configuration has the four-sublattice structure, where the spin vectors point the vertices of a tetrahedron if we put their bottoms at its center, as shown in Fig. 2(b). Hence we call this spin structure as "tetrahedral" structure. In fact we can prove in the same way that the tetrahedral structure is the ground state at least in the parameter region $-K/2 \leq J \leq 2K$.

For general values of J and K we determine the ground state in the mean-field approximation considering both three- and four-sublattice spin structures. In a three-sublattice structure with the sublattices a , b , and c , the energy is expressed as

$$\begin{aligned} E/N = & (J + 4K)(\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_b + \boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_c + \boldsymbol{\sigma}_c \cdot \boldsymbol{\sigma}_a) \\ & + 2K\{(\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_b)(\boldsymbol{\sigma}_c \cdot \boldsymbol{\sigma}_a) + (\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_c)(\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_b) \\ & + (\boldsymbol{\sigma}_c \cdot \boldsymbol{\sigma}_a)(\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_c)\} + 3K. \end{aligned} \quad (6)$$

In a four-sublattice structure with the sublattices a , b , c and d , we have

$$\begin{aligned} E/N = & \frac{1}{2}(J + 6K) \sum_{\alpha < \beta} \boldsymbol{\sigma}_\alpha \cdot \boldsymbol{\sigma}_\beta \\ & + K\{(\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_b)(\boldsymbol{\sigma}_c \cdot \boldsymbol{\sigma}_d) + (\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_c)(\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_d) \\ & + (\boldsymbol{\sigma}_a \cdot \boldsymbol{\sigma}_d)(\boldsymbol{\sigma}_b \cdot \boldsymbol{\sigma}_c)\}. \end{aligned} \quad (7)$$

In above equations $\boldsymbol{\sigma}_a$ denotes the spin vectors on the sublattice a etc. We determined the ground state spin configuration by comparing the energies of the two configurations which minimize eqs. (6) and (7), respectively. Minimization of eqs. (6) and (7) were done numerically. As a result we obtained four ground state phases with following spin structures:

1. the 120° structure with $E/N = -3(J + K)/2$ for $0 < K < 3J/25$,

2. the tetrahedral structure with $E/N = -J - 17K/3$ for $0 < 3J/25 < K$ and $0 < -3J/8 < K$,
3. the *uuud* structure with $E/N = -3K$ for $0 < -J/8 < K < -3K/8$,
4. the perfect ferromagnetism with $E/N = 3J + 21K$ for $0 < K < -J/8$.

The *uuud* structure has four sublattices and spins on the three sublattices are aligned parallel while spins on the other one is antiparallel to them. The ground-state spin structures are depicted in Fig. 2.

The most interesting and novel one among the obtained ground-state spin structures is the tetrahedral structure. It is remarkable that this structure has a scalar chiral long-range order. The scalar chirality in the unit cell may be defined as

$$\chi_S = \boldsymbol{\sigma}_a \cdot (\boldsymbol{\sigma}_b \times \boldsymbol{\sigma}_c) + \boldsymbol{\sigma}_b \cdot (\boldsymbol{\sigma}_d \times \boldsymbol{\sigma}_c) + \boldsymbol{\sigma}_c \cdot (\boldsymbol{\sigma}_d \times \boldsymbol{\sigma}_a) + \boldsymbol{\sigma}_d \cdot (\boldsymbol{\sigma}_b \times \boldsymbol{\sigma}_a), \quad (8)$$

which is an order parameter of Ising-type with a discrete Z_2 symmetry. Since the tetrahedral ground state has a finite value of χ_S , $16\sqrt{3}/9$, the system may undergo a phase transition of Ising universality at a finite temperature in spite of the isotropic rotational symmetry inherent in the Hamiltonian. This contrasts with the 120° structure which has a vector chiral order and may cause only topological phase transitions at finite temperatures.¹⁶

III. PHASE DIAGRAM IN THE MAGNETIC FIELD

Application of a magnetic field to frustrated spin systems is known to cause interesting magnetization processes and/or various phase transitions. First we consider the case with $K = 0$ and $J > 0$, i.e. the HAF on the triangular lattice with nearest neighbor coupling J . It is well known that two phase transitions occur under the magnetic field at $T=0$.¹⁴ When the magnitude of the field H is less than $3J$ (the magnetic moment of a spin is assumed as unity), The ground state is the so-called umbrella state with a coplanar spin configuration where the spins on a sublattice orient downwards (the magnetic field is assumed to be applied upwards) and those on the other two sublattices orient upwards but canted to the field by an angle (see Fig. 3(a)). The angle decreases with the magnetic field and vanishes at $H = 3J$. For $3J < H < 9J$, spins on all sublattices are canted to the field but two of them are parallel to each other (see Fig. 3(c)). We call the phase with this spin structure as c_3 phase. Spins are perfectly polarized for $H > 9J$. At $H = 3J$ the spins are collinear, i.e. those on two sublattices are oriented upwards while those on the other sublattice point downwards (see Fig. 3(b)). We call this spin structure as *uud* structure. It was shown that the phase with the *uud* structure expands and is realized in

a finite range of the magnetic field aided by the thermal or the quantum mechanical fluctuations.^{14,15}

We searched for the ground state for general values of J and K numerically minimizing eqs. (6) and (7) added by $-(\sigma_a^z + \sigma_b^z + \sigma_c^z)H/3$ and $-(\sigma_a^z + \sigma_b^z + \sigma_c^z + \sigma_d^z)H/4$, respectively. We found seven ground-state phases in total. They are the following:

1. the umbrella phase, which appears for a weak magnetic field when $J > 0$ and K is small.
2. the *uud* phase which extends around $H \simeq 3J(> 0)$ and for small K .
3. the c_3 phase, which appears for a wide range of the parameters both for positive and negative J near the boundary of the fully polarized phase.
4. the weak field four-sublattice phase, the spin structure is an analogue of the umbrella structure but is not coplanar. The spins on a sublattice point downwards and those on the other three sublattices orient upwards but canted to the field by an angle (see Fig. 3(d)). There is a three-fold rotational symmetry about the magnetic field axis. We call this structure "mushroom" structure. This phase appears for large K and small H .
5. the *uuud* phase which appears for small $|J|$ and rather weak field.
6. the high field four-sublattice phase with the structure analogous to the c_3 phase. Spins on three sublattices are parallel to each other and all spins are canted to the field (see Fig. 3(e)). We call this phase as c_4 phase.
7. the ferromagnetic phase where the spins are perfectly polarized. This phase appears for $H \geq 9(J + 8K)$.

Above spin structures are shown in Fig. 3. The phase diagram is depicted in Fig. 4 for $J > 0$ and in Fig. 5 for $J < 0$. We adopted a scale as $J + 8K = 1$ for $J > 0$ and $-2J + 8K = 1$ for $J < 0$ for convenience. The full lines denote the first order transitions while dotted lines the second order ones. The transitions between the three- and the four-sublattice structures are necessarily of the first order. The transitions between the structures with the same sublattice structures are of the second order in most cases. An exception is the boundary between the mushroom and the *uuud* phases for $J < 0$, where the phase change occurs with a magnetization jump. The critical field H_C at the second order transition points are determined analytically and tabulated in table 1.

There is a tetracritical point at $H_C = 6J = 24K$ when $J = 4K$. Magnetization process may show peculiar behaviors around this point. As an example magnetization per spin m is shown in Fig. 6 as a function of the applied field for $J = 0.4$ and $K = 0.75$. In this case the system undergoes six phase transitions with variation of H .

There appear two plateaus in the magnetization curve at $m = 1/3$ and $m = 1/2$. The $1/3$ - and $1/2$ -plateaus correspond to the *uud* and *uuud* phases, respectively. Furthermore the curve shows magnetization jumps at the first-order phase-transition points. Another magnetization curve is shown in Fig. 7 for the parameter $J = -0.1$ and $K = 0.1$.

IV. SUMMARY AND DISCUSSION

We have shown that various ground-state phases occur on the triangular lattice by coexistence of the two- and the four-spin exchange interactions. We have found new phases with four sublattices, i.e. the tetrahedral structure and the *uuud* structure. Since we have assumed that the ground states have either the three- or the four-sublattice structure, there remains a possibility that a state with a longer period may be more stable than those obtained above. In some cases, however, we are certain that the present results are rigorous in the classical limit. That is, the case when $K = 0$ (the pure Heisenberg model), and the case when $J = 0$ and $H = 0$. So it is quite reasonable that the obtained phases are stable in the regions close to these limiting cases. In fact we could prove that the tetrahedral phase is stable for small $|J|$.

It is remarkable that the phase with scalar chirality appears when the four-spin exchange dominates. This suggests the existence of a phase transition of the Ising universality at a finite temperature. Our preliminary Monte Carlo simulations in the classical limit clearly exhibit divergence of the specific heat at $T \simeq K$ for $J = 0$. The present model seems to be the first example of a realistic spin system in two dimensions with full rotational symmetry which undergoes a chiral phase transition. Detailed analysis of the finite-temperature properties will be reported elsewhere.¹⁷

In the region where the competition between the two- and the four-spin interactions are serious, more study is necessary to be conclusive. In fact we found in Monte Carlo simulations that various states with long periods and zero magnetization have quite close energy to that of the *uuud* structure. Furthermore, it can be shown that a new 120° structure state with nine sublattices has the same energy as the *uuud* structure. This indicates that the frustration of the system is strong in the phase with *uuud* structure due to the competition between the two- and the four-spin interactions. The application of the magnetic field would select the *uuud* structure from the degenerate ground states, since it has the largest magnetization among them. We hence believe that the ground-state phase diagram under the magnetic field (Fig. 5) will not be changed seriously by the appearance of long-period states.

Effects of quantum fluctuations, which we have completely neglected in the present study, are surely very

important since the magnitude of the nuclear spin is $1/2$ for ${}^3\text{He}$. It is still not quite established that even the well-known 120° structure is stable against quantum fluctuations when $K = 0$ and $J > 0$ (HAF), though recent numerical studies seem to support for the stability.¹⁸ It is possible that the coexistence of the two- and the four-spin interactions destabilizes the ordered ground state and helps the quantum fluctuations to destroy it. The stability of the ground states against the zero-point spin wave fluctuations are presently under investigation and will be reported elsewhere.¹⁹

Finally we compare our results with the existing experimental ones, though it is still not quite clear whether the model is relevant to describe ${}^3\text{He}$ layers. The specific heat data at low temperatures exist only at two different values of the coverage, i.e. $\rho = 0.180 \text{ \AA}^{-2}$ and 0.228 \AA^{-2} .^{7,20} No sharp peak was observed at both coverages. The system with $\rho = 0.228 \text{ \AA}^{-2}$ clearly corresponds to a negative J since it shows about 60 percent of full polarization under a very small magnetic field.²¹ On the other hand the parameter J of the system with $\rho = 0.180 \text{ \AA}^{-2}$ is still unclear. At this coverage the system has been thought to be antiferromagnetic since it has a negative Weiss temperature θ . However a negative θ does not imply a positive J since $\theta = -6(J + 6K)$ holds in the present model. If the system corresponds to a positive J , the ground state at this coverage should be the positive large- J phase or a new quantum phase caused by competitions between the two- and the four-spin interactions and quantum fluctuations, since the system does not show a finite-temperature phase transition. If this is the case, there may occur the phase with a chiral long-range order between two coverages $\rho = 0.180 \text{ \AA}^{-2}$ and 0.228 \AA^{-2} (if the density of the second layer varies continuously), and we may observe a sharp peak in the specific heat by varying the coverage of the adsorbed layers. Another possibility is that the system with $\rho = 0.180 \text{ \AA}^{-2}$ has a negative J whose value corresponds to the *uuud* ground state in the mean field theory. In this parameter region we have seen that the strong frustration leads to the existence of various states with long periods whose energies are quite close to that of the ground state. Then the effects of quantum fluctuations may realize a novel ground state without magnetic long-range order. Observations under a magnetic field will be useful to determine the phase that the system belongs to. Above discussion is based on the assumption that more-than-four-spin exchange interactions are not important. If this is not the case it is necessary to investigate a spin model with more-than-four-spin exchange interactions in search for possible ground states.

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Table

Phase1	Phase2	H_C
umbrella	<i>uud</i>	$3(J - 4K)$
<i>uud</i>	c_3	$3(J + 4K)$
ferromagnetic	c_3	$9(J + 8K)$
mushroom	<i>uuud</i>	$4(J + 2K)$
<i>uuud</i>	c_4	$4(J + 8K)$

TABLE I. H_C at the second order transition point.

Figure Captions

FIG. 1. (a) A unit diamond described by the four-spin exchange Hamiltonian (2). (b) A unit hexagon described by H_h in eq. (5).

FIG. 2. The ground state spin structures: (a) the 120° , (b) the tetrahedral and (c) the *uuud* structure. The ferromagnetic spin structure is an obvious one.

FIG. 3. The ground state spin structures in the magnetic field: (a) the umbrella, (b) the *uud*, (c) the c_3 , (d) the mushroom and (e) the c_4 structure. The *uuud* structure is shown in Fig. 2. The magnetic field is assumed to be applied upwards.

FIG. 4. The ground state phase diagram for $J > 0$. The parameters J and K are scaled as $J + 8K = 1$. The phases are labelled by u(umbrella), m(mushroom) and f(perfect ferromagnetism). Other labels are self-evident. The full and the dotted lines indicate the phase boundaries of the first and the second orders, respectively.

FIG. 5. The ground state phase diagram for $J < 0$. The parameters J and K are scaled as $-2J + 8K = 1$. The labels and the lines are the same as in Fig. 4.

FIG. 6. The magnetization curve for $J = 0.4$ and $K = 0.75$. The system undergoes three first-order transitions and three second-order ones. The c_3 phase appears twice.

FIG. 7. The magnetization curve for $J = -0.1$ and $K = 0.1$. The system undergoes two first-order transitions and two second-order ones.